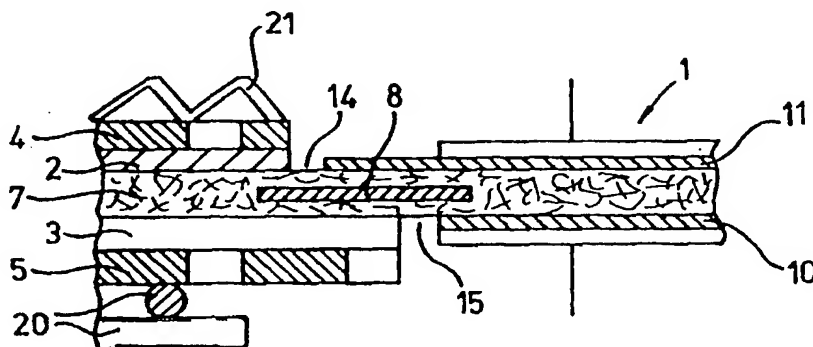




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<p>(21) International Application Number: PCT/NL99/00271</p> <p>(22) International Filing Date: 4 May 1999 (04.05.99)</p> <p>(30) Priority Data: 1009062 4 May 1998 (04.05.98) NL</p> <p>(71) Applicant (for all designated States except US): STICHTING ENERGIEONDERZOEK CENTRUM NEDERLAND [NL/NL]; Westerduinweg 3, NL-1755 LE Petten (NL).</p> <p>(72) Inventor; and (75) Inventor/Applicant (for US only): RIETVELD, Gijsbertus [NL/NL]; Bohemenstraat 8, NL-1827 EH Alkmaar (NL).</p> <p>(74) Agent: DE BRUIJN, Leendert, C.; Nederlandsch Octrooibureau, Scheveningseweg 82, P.O. Box 29720, NL-2502 LS The Hague (NL).</p>	<p>(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).</p> <p>Published With international search report. In English translation (filed in Dutch).</p>	

(54) Title: SEAL FOR MATRIX PLATE ELECTRODE



(57) Abstract

Electrochemical cell consisting of a sealing matrix plate on which electrode (2, 3) assemblies abut on either side. The matrix plate is provided with openings for supplying/discharging gases participating in the electrochemical reaction. Close to said openings there are support plates (10, 11) which support the electrodes. In order to prevent local leakage, it is proposed to arrange foil material in or adjoining the matrix plate (7), which foil material extends over said transition. Moreover, it is possible for the end delimiters of the cathode (2) and anode (3) to have been arranged offset with respect to one another with respect to a perpendicular to the matrix plate.

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Seal for matrix plate electrode

The present invention relates to an electrochemical cell according to the preamble of Claim 1.

5 A cell of this type is disclosed in US Patent 5 298 342. In MCFC cells the matrix plate consists of a ceramic material that has been filled with a carbonate material solidified at room temperature. On heating to approximately 650 °C the carbonate becomes fluid and provides, on the one hand, the electrolyte and, on the other hand, a seal between the electrodes.

A matrix plate of this type is provided with openings for transporting gases in a
10 direction perpendicular to the plate. Close to said openings there is a special construction on either side of the matrix plates, which special construction provides for communication between such an opening and the gas distribution element of the electrode concerned. A special construction of this type must be constructed such that a seal is provided with respect to the other electrode, that is to say the electrode that is located on the other side of the
15 matrix. To this end support plates are used which meet the separator plate and are joined to one another, so as to provide a seal, at the location of the opening. By way of example a stainless steel support plate is used. These support plates can be provided with resilient devices to ensure sealing. A construction of this type is described in EP 0 408 104 A1 in the name of ECN, Petten, the Netherlands.

20 According to a further proposal, the support plates can be provided with elevations or ribs which are of a highly local nature and in combination with the matrix plate prevent cracking of the latter. Such cracks can arise during operation and especially during warming up. Such cracks form openings extending through the matrix and, certainly in the region between support plate and electrode, which region will hereinafter be referred to as transition,
25 can have particularly disastrous effects because in an extreme case the reactants can come into direct contact with one another.

It has been found that despite the abovementioned measures the transition region is particularly critical. After all, on one side of the transition region the matrix plate is under pressure from the support plate, whilst on the other side of the transition pressure exerted on
30 the electrode provides sealing force. Pressure is exerted on the electrode, in turn, by the gas distribution assembly, which optionally bears directly on the separator plate. It has been found that local stresses can arise as a result of this differing construction of support plate and electrode assembly and as a result of firing up the matrix, as a result of which stresses

cracking occurs mainly close to said transition.

In US Patent 5 298 342 the matrix plate is separated into two parts and a foil is arranged between these. At the location of the actual electrochemical cell the foil is provided with openings, so that the electrochemical reaction can take place. The foil is of sealed construction close to the transitions. Because the foil extends over the entire length of the matrix, the thickness of the electrochemical cell will increase by the thickness of the foil component. It has been observed that, despite the improved sealing by the foil component, the matrix material still cracks close to the transition.

The aim of the present application is to restrict the consequences of such cracking as far as possible.

This aim is achieved with an electrochemical cell as described above by means of the characterising measures in Claim 1. By applying the foil material only locally, that is to say not extending over the entire cell surface, as in the abovementioned US Patent 5 298 342, a higher pressure can be generated locally. As a result a certain degree of compression is produced in the matrix and, surprisingly, it has been observed that as a result cracking in the matrix is appreciably less or completely excluded.

In general, such compression is achieved if at least 50 % of the thickness of the foil component is taken up in the surrounding matrix material by compression of the latter. According to a preferred embodiment of the invention, however, the compression that takes place is such that the matrix at the location of the transition, that is to say including the foil component located therein, is no more than 30 % thicker than the rest of the matrix.

A local foil layer of this type can be applied in various ways. For instance, it is possible for the foil layer to have been applied such that it extends parallel to the boundary surface of the matrix plate, the layer having been placed against one side of the matrix plate and contact pressure being thus exerted thereon by the support plate and the electrode concerned. It is also possible to make a recess in the matrix plate or to make said plate of stepped construction. On the other hand, such a recess will automatically be formed during heating of the electrochemical cell. It is also possible to allow such a foil layer to extend from the heart of the matrix to the edge limit. This can be of importance when a metallic foil layer is used where direct electrical contact between support plate and electrode and foil is not desired or, on the contrary, is desired. Of course, numerous other embodiments of the foil layer are possible, sealing always being provided in the event of cracking of the matrix plate.

According to an advantageous embodiment of the invention, the foil material consists

of a stainless steel support on which an aluminium layer has been applied. During warming up of the fuel cell to the operating temperature the aluminium layer reacts with the support and an aluminising layer is formed which consists of a mixture of intermetallic compounds of, on the one hand, iron, nickel and chromium and, on the other hand, aluminium. This
5 aluminising layer has a melting point that is significantly higher than the operating temperature of the cell. During operation of the cell the aluminium in the aluminising layer reacts with the carbonate material to form a sealing layer (in general this will be lithium aluminate) which counteracts further attack. It is, of course, also possible to pre-treat the aluminium-coated stainless steel component in a furnace. However, it has been found that the
10 mechanical properties become poorer as a result of the formation of such intermetallic compounds, as a consequence of which fracture can easily occur on subsequent fitting in the cell. Therefore preference is given to incorporating the aluminium-coated stainless steel as produced in the cell, the corrosion resistance being obtained during start-up of the cell. Instead of stainless steel it is possible to use any other ferrous metal or nickel alloy. It has
15 been found that the life of such a foil material at least corresponds to the life of a fuel cell, that is to say at least 50,000 hours under the adverse conditions which prevail in such a fuel cell with molten carbonate.

The thickness of the foil material is preferably between 0.05 and 0.6 mm and more particularly is approximately 0.2 mm. In the case of an abovementioned foil component
20 consisting of stainless steel and an aluminium layer, the aluminium layer is present in a thickness of between 10 and 60 μm and preferably 20 μm . The aluminium is applied, for example, by PVD.

As indicated above, the foil material can comprise a metallic material. This will be the case in particular if operating at elevated temperatures, as in an MCFC cell. However, it is
25 certainly conceivable to use foil layers in electrochemical cells which operate at relatively low temperatures. In this case plastic materials can be used. If metallic material is used it is, of course, important that the material is not (too rapidly) attacked by chemical reactions and/or does not prevent the electrochemical reaction. To this end the foil material is preferably a (semi-)noble material such as copper, nickel, silver and/or gold or a material having a high
30 corrosion resistance, such as aluminium-alloyed nickel or steel, or an aluminised metal.

With a standard embodiment of an electrochemical cell the anode-support plate end delimitation and the cathode-support plate end delimitation are in line with one another with respect to a perpendicular to the plane of the matrix. It has been found that as a result local

high stress concentrations can arise in the matrix, which is one of the reasons for cracking, the effect of which can be eliminated by means of the measures described above.

Such cracking can be appreciably restricted or prevented by no longer arranging the anode-support plate/cathode-support plate end delimitations in one line with respect to a perpendicular to the separator plate. That is to say the end delimitations are offset with respect to one another. The adaptability of the construction can be increased by making one or both gas distribution elements again offset with respect to the electrode current collector assembly.

The invention will be explained in more detail below with reference to an illustrative embodiment shown in the drawings. In the drawings:

- Fig. 1 shows part of a fuel cell stack close to a discharge opening for gases;
- Fig. 2 shows a detail from Fig. 1 in a first embodiment;
- Fig. 3 shows a second embodiment of the detail shown in Fig. 1, and
- Fig. 4 shows, in detail, a modified foil component according to the invention.

In Fig. 1 the fuel cell stack according to the invention is indicated in its entirety by 1. This fuel cell stack consists of a number of fuel cells 13. Each fuel cell 13 consists of a cathode 2 and an anode 3. In this example a so-called MCFC (molten carbonate fuel cell) is shown. A cathode current collector, which is indicated by 4, is joined to the cathode 2 and the same applies in respect of the anode 3, which is joined to an anode current collector 5. Gas distribution elements about the current collectors. The gas distribution elements are indicated by 21 for the cathode and by 20 for the anode. The gas distribution element for the anode consists, for example, of a wire mesh.

A separator plate is indicated by 6, whilst the matrix plate containing carbonate material is indicated by 7. Such a plate can be produced by any method known from the prior art, such as by tape casting. Only the discharge of gas via element 9 is shown in the drawing.

Two support plates join onto the separator plate 6. Said support plates are indicated in Fig. 2 by 10 and 11. These plates are stainless steel components which have been welded to the separator plate 6 close to opening 9 so as to provide a seal. Openings, which are not indicated in more detail, will be present in one of the support plates in order to provide gas communication with either the cathode gas distribution element or the anode gas distribution element. As can be seen from Fig. 2, the support plates 10, 11 extend over a relatively short length and then merge into the anode 3 and, respectively, the cathode 2. There is a transition 14 between the end delimitation of support plate 11 and cathode 2. The transition between anode 3 and support plate 10 is indicated by 15. It can be seen from the drawing that these

have been arranged offset with respect to one another. In the transition area, that is to say close to transitions 14 and 15, a foil 8 of metallic material has been arranged in the heart of the matrix 7. The foil extends essentially parallel to the plane of the matrix plate 7. On the anode side the gas distribution element 20 extends less far than the corrugation 5. The length of the foil component 8 is indicated by 1 in Fig. 2. According to the invention this length is between 5 and 21 mm and is preferably approximately 9 mm. In the latter case always 3 mm of the foil extends in a different area of the electrochemical cell, that is to say 3 mm extends beyond component 11, 3 mm extends in the area where only component 11 and not component 10 as well is located and 3 mm extends in an area where both component 11 and component 10 are present.

In order to apply such a foil layer, it is possible to make up the matrix from three layers. With this arrangement the middle layer is locally interrupted and the foil layer is applied or a cavity is produced by compressing the matrix during start-up/operation at the location of the foil.

It is, of course, possible for said foil to be arranged closer to transition 14 or transition 15. It is even possible for said foil to have been applied such that it is in contact with either support plate 10 or support plate 11. There is optionally contact with cathode 2 or anode 3.

Because a metallic foil has its own corrosion potential, the position thereof is of importance. Fewest problems are found to occur with positioning in contact with the anode. After all, the anode potential is more negative than the cathode potential. As a result of the offset transitions 14 and 15, the risk of the occurrence of fracture in the matrix, which risk is real during the warm-up phase of, for example, an MCFC cell from the green state to approximately 650 °C, is appreciably reduced, despite the appreciable stress concentration close to the transitions. The gas leakage that is caused by fracture is completely prevented by the presence of foil. The foil material can be any material that is able to withstand the environment in the matrix and in the case of an MCFC cell this will be carbonate material that is appreciably aggressive at 650 °C, so that preference is given to a (semi-)noble metal foil.

By way of example, a foil of this type, for example made of gold material, can have a thickness of 50 µm. Instead of (semi-)noble metals, other materials able to withstand the environment concerned can also be used. A nickel foil containing 5 - 50 % by weight aluminium may be mentioned by way of example.

Another possibility is a stainless steel alloy containing an appreciable proportion of

aluminium.

As can be seen from Fig. 3, variants of the invention are readily conceivable. In the embodiment according to Fig. 3, all components which correspond to those shown in Fig. 2 have been provided with the same reference numerals. The transitions 16 and 17 are now directly opposite one another, so that an appreciable stress concentration will be produced. Gas distribution element 20 has the same length, whilst element 21 has been offset somewhat. The foil layer is indicated here by 18 and it can be seen that said layer is in contact with anode 3. So as to avoid contact with support plate 10, said foil layer extends into the interior of the matrix close to said support plate 10.

In Fig. 4 a further foil component is indicated diagrammatically by 28. This can be used either as a curved foil component or as a straight foil component. This consists of a core of stainless material of the 300 series, which core is indicated by 29, provided with a layer of aluminium applied thereon on all sides, which layer of aluminium is indicated by 30. The thickness of the layer of stainless steel is 0.2 mm, whilst the aluminium layer applied by PVD has a thickness of 20 μm . Such a strip of material is arranged in the electrochemical cell and on starting up the electrochemical cell, under the influence of high temperature and the environment prevailing in the cell, an aluminate is formed by aluminium material that forms a compound with the lithium or potassium present in the matrix. Moreover, an intermetallic aluminium-nickel-chromium-iron compound is formed.

It will be understood that numerous variants for the positioning of the foil layer are possible. The only essential aspect is that, by local compression of the matrix, the foil prevents cracking in said matrix and, in the unlikely event of fracture still occurring, that passage of gases and the coming together of reactants is effectively prevented. It will be understood that such variants lie within the scope of the appended claims.

Claims

1. Electrochemical cell (13) comprising an electrolyte matrix (7) which provides a seal in operation and is delimited on either side by electrode assemblies, each comprising an electrode (2, 3) and a corrugation/gas distribution device (4, 5) enclosed between separator plates (6), wherein said matrix is provided with openings (9) for supplying/discharging gases in a direction perpendicular to said matrix plate, wherein, in the region directly adjoining said opening, the matrix is supported by a support plate (10, 11), the electrode concerned abutting said support plate, wherein a foil component (8, 18, 28) which seals the transition (14, 15, 16, 17) between support plate and electrode is arranged at said transition, characterised in that said foil component (8, 18, 28) extends exclusively close to said transition and wherein the total thickness of the matrix and the foil component close to said transition is essentially less than 50 % of the foil thickness greater than the total thickness of the matrix elsewhere in said cell.
2. Electrochemical cell according to Claim 1, wherein the length (1) of the foil component is between 5 and 21 mm.
3. Electrochemical cell according to one of the preceding claims, having a thickness of between 0.05 and 0.6 mm.
4. Electrochemical cell according to one of the preceding claims, wherein said foil component is arranged at least partially in contact with the matrix plate and between the support plate and electrode.
5. Electrochemical cell according to one of the preceding claims, wherein said foil component is arranged at least partially in the matrix plate.
6. Electrochemical cell according to one of the preceding claims, wherein said foil component comprises a metallic material.
7. Electrochemical cell according to Claim 4, wherein said metallic material comprises a (semi-)noble metal.
8. Electrochemical cell according to Claim 4, wherein said metallic material comprises an aluminium-containing metal.
9. Electrochemical cell according to one of the preceding claims, wherein said foil component (28) comprises a stainless material coated with an aluminium-containing layer.
10. Electrochemical cell according to Claim 9, wherein said stainless material comprises stainless steel.

11. Electrochemical cell according to one of the preceding claims, wherein an electrode/support plate transition is arranged on either side of the matrix plate, wherein said transitions (14, 15, 16, 17) are arranged offset with respect to one another with respect to a perpendicular to the matrix plate.

- 5 12. Electrochemical cell according to Claim 11, wherein the end delimitations of the gas distribution elements of the opposing electrode assemblies are offset with respect to electrode and current collector.

fig-1

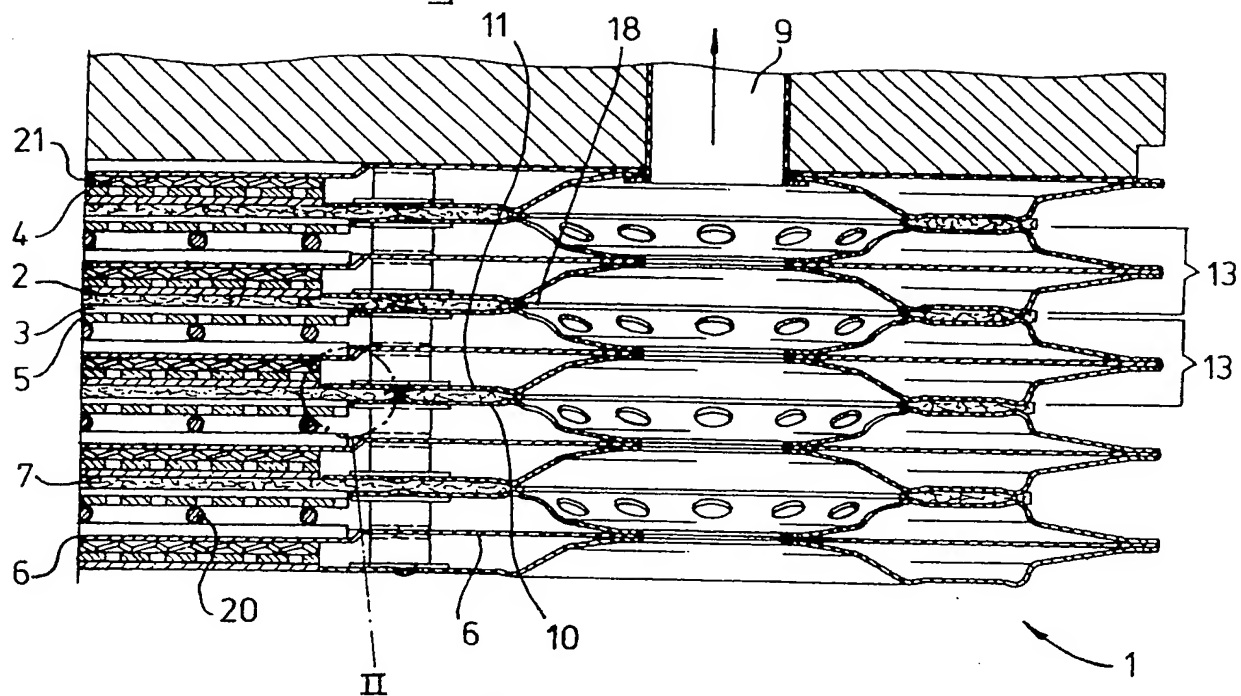


fig - 2

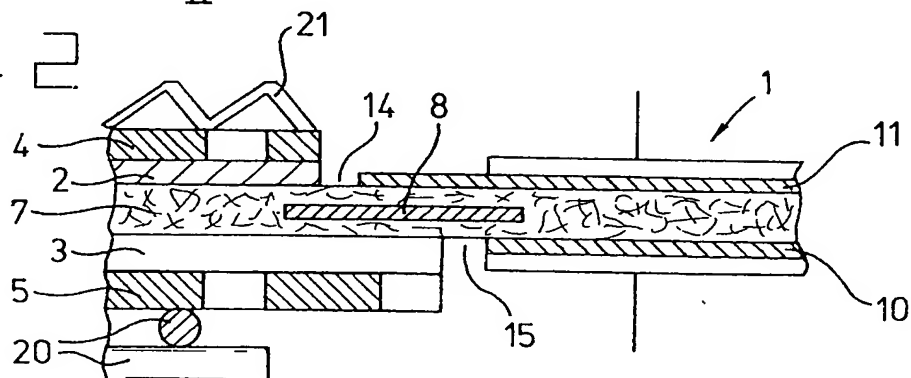


fig-3

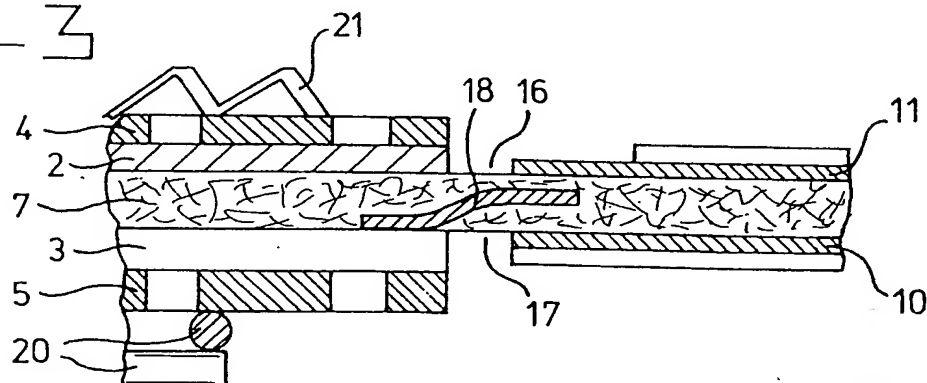
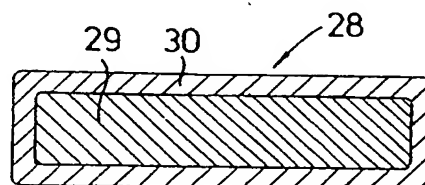


fig-4



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C. DOCUMENTS CONSIDERED TO BE RELEVANT

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Date of the actual completion of the international search

10 August 1999

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INTERNATIONAL SEARCH REPORT

International Application No

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International Application No

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